

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Application of:	Rath	Docket No.:	49091
Serial No.:	09/701,587	Confirmation No.:	5855
Filing Date:	11/30/2000	Examiner:	LU, C CAIXIA
Customer No.:	26474	Art Unit:	1713

For: Method for producing highly reactive polyisobutenes

Honorable Commissioner for Patents  
P.O. Box 1450  
Alexandria, Virginia 22313-1450

Appeal brief under 37 C.F.R. § 41.37

Sir:

This is an appeal from the Examiner's final rejection of Claims 12, 14 – 17 and 21 – 35, dated December 14, 2006. Claims 12, 14 – 17 and 21 – 35 are currently pending.

It is respectfully requested that a two month extension of time be granted in this case. The respective \$450.00 fee is paid by credit card (Form PTO-2038 enclosed).

The fee set forth in 37 C.F.R. § 41.20(b)(2) is paid by credit card. Form PTO-2038 is enclosed. Please charge any shortage in fees due in connection with the filing of this paper, including Extension of Time fees, to Deposit Account 14.1437. Please credit any excess fees to such account.

Respectfully submitted,  
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Real party in interest:

The real party in interest is BASF Aktiengesellschaft, of Ludwigshafen, Germany.

Related appeals and interferences:

To the best of the undersigned's knowledge, there are no related interferences or judicial proceedings.

Status of claims:

- Claims 12, 14 – 17 and 21 – 35 are pending in the application
- Claims 12, 14 – 17 and 21 – 35 stand rejected and are being appealed.
- Claims 1 – 11, 13, 18 – 20 are canceled.

Status of amendment:

No amendment was filed subsequent to the final rejection dated December 14, 2006.

Summary of claimed subject matter:

The claimed invention relates to the preparation of certain highly reactive polyisobutenes. The independent claims involved in the appeal are claims 12 and 27. Claims 14 – 17 and 21 – 26, 34 and 35 depend either directly or indirectly from claim 12. Claims 28 – 33 depend from claim 27.

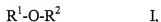
Independent claim 12 relates to a process for preparing highly reactive polyisobutenes by cationic polymerization of isobutene in the liquid phase<sup>1</sup> in the presence of a complex comprising boron trifluoride<sup>2</sup> and a) a primary alcohol having 1-

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<sup>1</sup> Specification, page 1, line 8.

<sup>2</sup> Specification, page 1, lines 8 – 9.

20 carbon atoms or a secondary alcohol having 3-20 carbon atoms, or a mixture of these alcohols,<sup>3</sup> and b) an ether containing no tertiary alkyl groups and having the formula I,<sup>4</sup>



wherein R<sup>1</sup> is a secondary alkyl group having 3-10 carbon atoms<sup>5</sup>, and R<sup>2</sup> is methyl, ethyl, or a primary or secondary alkyl group having 3-10 carbon atoms,<sup>6</sup> wherein the alcohol (a) and the ether (b) have a molar ratio of from 0.01:1 to 1:1,<sup>7</sup> and wherein the molar ratio of the sum of alcohol (a) and ether (b) to boron trifluoride is from 1.4:1 to 2:1.<sup>8</sup> The process is carried out at from +40° to -60°C.<sup>9</sup> The highly reactive polyisobutenes prepared by the claimed process have a terminal vinylidene group content of more than 80 mol%<sup>10</sup> and a number average molecular weight of from 500 to 5000 dalton.<sup>11</sup>

Claim 25, which depends from claim 12 relates to the process as claimed in claim 12, wherein polyisobutenes having a terminal vinylidene group content of more than 90 mol%<sup>12</sup> are polymerized at an isobutene conversion of up to 95%<sup>13</sup> using a preformed complex of boron trifluoride/isopropanol/diisopropyl ether,<sup>14</sup> having a molar ratio of isopropanol to diisopropyl ether of from 2:1 to 1:5<sup>15</sup> and a molar ratio of boron trifluoride to diisopropyl ether of from 0.6:1 to 0.9:1.<sup>16</sup>

Independent claim 27 is a product-by-process claim directed to a polyisobutene having a number average molecular weight of from 500 to 5000 dalton<sup>17</sup> and a terminal vinylidene group content of more than 90%,<sup>18</sup> obtained by cationic polymerization of isobutene in the liquid phase<sup>19</sup> with the aid of boron trifluoride as catalyst at from 40 to -

<sup>3</sup> Specification, page 5, lines 36 – 38.

<sup>4</sup> Specification, page 5, lines 40 – 43.

<sup>5</sup> Specification, page 6, lines 1 – 2.

<sup>6</sup> Specification, page 6, lines 1 – 2.

<sup>7</sup> Specification, page 9, lines 20 – 22.

<sup>8</sup> Specification, page 9, lines 29 – 31.

<sup>9</sup> Specification, page 1, line 9.

<sup>10</sup> Specification, page 1, lines 5 – 6.

<sup>11</sup> Specification, page 1, lines 6 – 7.

<sup>12</sup> Specification, page 6, lines 38 – 39.

<sup>13</sup> Specification, page 4, lines 8 – 9.

<sup>14</sup> Specification, page 7, line 37; and page 13, line 25 (example 1).

<sup>15</sup> Specification, page 9, line 22.

<sup>16</sup> Specification, page 16, lines 9 – 10.

<sup>17</sup> Specification, page 1, lines 6 – 7.

<sup>18</sup> Specification, page 6, line 39.

<sup>19</sup> Specification, page 1, line 8.

60°C<sup>20</sup> in the presence of a boron trifluoride complex<sup>21</sup> with a) a primary alcohol having 1-20 carbon atoms or a secondary alcohol having 3-20 carbon atoms, or a mixture of these alcohols,<sup>22</sup> and b) an ether containing no tertiary alkyl groups and having the formula I,<sup>23</sup>



wherein R<sup>1</sup> is a secondary alkyl group having 3-10 carbon atoms,<sup>24</sup> and R<sup>2</sup> is methyl, ethyl, or a primary or secondary alkyl group having 3-10 carbon atoms,<sup>25</sup> wherein the alcohol (a) and the ether (b) have a molar ratio of from 0.01:1 to 1:1,<sup>26</sup> and wherein the molar ratio of the sum of alcohol (a) and ether (b) to boron trifluoride is from 1.4:1 to 2:1.<sup>27</sup>

Grounds of rejection to be reviewed on appeal:

Claims 12, 14 – 17 and 21 – 35 under 35 U.S.C. §103(a) over Rath (US 5,408,018).

Argument:

In US 5,408,018, Rath teaches that the best results are achieved when the ether utilized contains at least one tertiary alkyl group.

Example 6 of Rath (US 5,408,018):

250 g/hr	isobutene,
250 g/hr	n-hexane,
20 mmol	boron trifluoride,
10 mmol	boron trifluoride/2 butanol complex.
21 mmol	<b>2-butyl tert butyl ether</b> (see figure below)

<sup>20</sup> Specification, page 1, line 9.

<sup>21</sup> Specification, page 1, lines 8 – 9.

<sup>22</sup> Specification, page 5, lines 36 – 38.

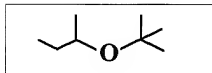
<sup>23</sup> Specification, page 5, lines 40 – 43.

<sup>24</sup> Specification, page 6, lines 1 – 2.

<sup>25</sup> Specification, page 6, lines 1 – 2.

<sup>26</sup> Specification, page 9, lines 20 – 22.

<sup>27</sup> Specification, page 9, lines 29 – 31.

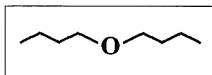
Results:

85%	isobutene conversion
1065	polyisobutene $M_n$
92%	terminal double bonds

When the ether utilized does not contain at least one tertiary alkyl group, Rath (US 5,408,018) teach that poorer results are achieved.

Example 8 of Rath (US 5,408,018):

250 g/hr	isobutene,
250 g/hr	n-hexane,
20 mmol	boron trifluoride,
10 mmol	boron trifluoride/2 butanol complex.
40 mmol	<b>di-n-butyl ether</b> (see figure below)

Results:

76%	isobutene conversion
990	polyisobutene $M_n$
80%	terminal double bonds

In US 5,408,018, Rath does not teach or suggest that by polymerizing isobutene in the presence of a complex comprising boron trifluoride and

- a primary alcohol having 1-20 carbon atoms or a secondary alcohol having 3-20 carbon atoms, or a mixture of these alcohols, and
- an ether containing no tertiary alkyl groups and having the formula I



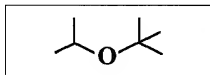
wherein  $R^1$  is a secondary alkyl group having 3-10 carbon atoms, and  $R^2$  is methyl, ethyl,

or a primary or secondary alkyl group having 3-10 carbon atoms, improved results are obtained.

Indeed, based on the disclosure of US 5,408,018, the results shown in the declaration under 37 C.F.R. §1.132 of Dr. Hans-Peter Rath, filed with the USPTO on January 16, 2003, are unexpected. In that declaration a process according to US 5,408,018 was compared to a process according to the claimed invention.

Comparative Example according to US 5,408,018:

300 g/hr	isobutene,
300 g/hr	hexane,
29.7 mmol/hr	boron trifluoride,
15 mmol/hr	isopropanol
25 mmol/hr	<b>isopropyl tert-butyl ether</b> (see figure below)

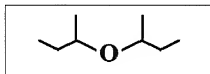


Results:

960 polyisobutene  $M_n$   
85.4% terminal double bonds

Example according to the present invention:

300 g/hr	isobutene,
300 g/hr	hexane,
21.4 mmol/hr	boron trifluoride,
10 mmol/hr	isopropanol
25 mmol/hr	<b>di-sec-butyl ether</b> (see figure below)



Results:

890 polyisobutene  $M_n$

95.9% terminal double bonds

The increase in the percentage of terminal double bonds (85.4% terminal double bonds in the comparative example according to US 5,408,018 versus 95.9% terminal double bonds in the example according to the present invention) upon polymerizing isobutene in the presence of a complex comprising boron trifluoride and

- a) a primary alcohol having 1-20 carbon atoms or a secondary alcohol having 3-20 carbon atoms, or a mixture of these alcohols, and
- b) an ether containing no tertiary alkyl groups and having the formula I



wherein R<sup>1</sup> is a secondary alkyl group having 3-10 carbon atoms, and R<sup>2</sup> is methyl, ethyl, or a primary or secondary alkyl group having 3-10 carbon atoms, is an unexpected result. "A greater than expected result is an evidentiary factor pertinent to the legal conclusion of obviousness ... of the claims at issue."<sup>28</sup>

The teaching of Rath contains nothing which could reasonably be taken to render applicant's invention as a whole *prima facie* obvious within the meaning of Section 103(a). In fact, the foregoing shows that the reference, when considered as a whole for what it reasonably taught to a person of ordinary skill at the time at which applicant made his invention, conveys that the utilization of ethers having at least one tertiary alkyl group is mandatory in order to arrive at polyisobutenes having a terminal vinylidene group content of more than 80%. It is well settled that the invention as a whole which is referenced in 35 U.S.C. 103(a) is not limited to the elements of features which are recited in a claim but also encompasses the properties which are achieved due to the particular combination of elements defined in the claim.<sup>29</sup> Since Rath illustrate that the use of ethers which lack a tertiary alkyl group results in a significant deterioration of the success in forming terminal vinylidene groups in the polyisobutenes a person of ordinary skill in the art could not reasonably expect that a process which does not employ the tertiary ethers

<sup>28</sup> In re Corkill, 711 F.2d 1496, 226 USPQ 1005 (Fed. Cir. 1985).

<sup>29</sup> In re Antonie, 559 F.2d 618, 620, 195 USPQ 6, 8 (CCPA 1977).

of Rath would yield in the highly reactive polyisobutenes referenced in applicants' claims.



Claims appendix.

1. - 11. (canceled)
12. (previously presented) A process for preparing highly reactive polyisobutenes having a terminal vinylidene group content of more than 80 mol% and a number average molecular weight of from 500 to 5000 dalton by cationic polymerization of isobutene in the liquid phase in the presence of a complex comprising boron trifluoride at from +40° to -60°C, which comprises polymerizing isobutene in the presence of a complex comprising boron trifluoride and
- a) a primary alcohol having 1-20 carbon atoms or a secondary alcohol having 3-20 carbon atoms, or a mixture of these alcohols, and
- b) an ether containing no tertiary alkyl groups and having the formula I



wherein  $R^1$  is a secondary alkyl group having 3-10 carbon atoms, and  $R^2$  is methyl, ethyl, or a primary or secondary alkyl group having 3-10 carbon atoms,

wherein the alcohol (a) and the ether (b) have a molar ratio of from 0.01:1 to 1:1, and

wherein the molar ratio of the sum of alcohol (a) and ether (b) to boron trifluoride is from 1.4:1 to 2:1.

13. (canceled)
14. (previously presented) A process as claimed in claim 12, wherein the molar ratio of the alcohol (a) to the ether (b) is from 0.2:1 to 1:1.
15. (previously presented) A process as claimed in claim 12, wherein the molar ratio of the alcohol (a) to the ether (b) is from 0.4:1 to 1:1.
16. (previously presented) A process as claimed in claim 15, wherein the alcohol (a) is isopropyl alcohol and/or 2-butanol.

17. (previously presented) A process as claimed in claim 15, wherein the ether (b) is diisopropyl ether, di-sec-butyl ether and/or isopropyl sec-butyl ether.
18. - 20. (canceled)
21. (previously presented) A process as claimed in claim 12, wherein the alcohol (a) is isopropyl alcohol and/or 2-butanol.
22. (previously presented) A process as claimed in claim 12, wherein the ether (b) is diisopropyl ether, di-sec-butyl ether and/or isopropyl sec-butyl ether.
23. (previously presented) A process as claimed in claim 12, wherein boron trifluoride, the alcohol (a) and the ether (b) are combined in the polymerization reactor to generate the complex in situ in the polymerization mixture.
24. (previously presented) A process as claimed in claim 12, wherein a complex of boron trifluoride and the ether (b) is preformulated and is introduced into the solvent or monomer feed to the reactor or directly into the reactor, either separately or together with the alcohol (a).
25. (previously presented) A process as claimed in claim 12, wherein polyisobutenes having a terminal vinylidene group content of more than 90 mol% are polymerized at an isobutene conversion of up to 95% using a preformed complex of boron trifluoride/isopropanol/diisopropyl ether, having a molar ratio of isopropanol to diisopropyl ether of from 2:1 to 1:5 and a molar ratio of boron trifluoride to diisopropyl ether of from 0.6:1 to 0.9:1.
26. (previously presented) A process as claimed in claim 12, wherein the isobutene source is a C<sub>4</sub> cut comprising isobutene in an amount of at least 6% by weight.
27. (previously presented) A polyisobutene having a number average molecular weight of from 500 to 5000 dalton and a terminal vinylidene group content of more than 90%, obtained by cationic polymerization of isobutene in the liquid phase with the aid of boron trifluoride as catalyst at from 40 to -60°C in the presence of a

boron trifluoride complex with

- a) a primary alcohol having 1-20 carbon atoms or a secondary alcohol having 3-20 carbon atoms, or a mixture of these alcohols, and
- b) an ether containing no tertiary alkyl groups and having the formula I



wherein  $R^1$  is a secondary alkyl group having 3-10 carbon atoms, and  $R^2$  is methyl, ethyl, or a primary or secondary alkyl group having 3-10 carbon atoms,

wherein the alcohol (a) and the ether (b) have a molar ratio of from 0.01:1 to 1:1, and

wherein the molar ratio of the sum of alcohol (a) and ether (b) to boron trifluoride is from 1.4:1 to 2:1.

- 28. (previously presented) A polyisobutene as claimed in claim 27, wherein the molar ratio of the alcohol (a) to the ether (b) is from 0.2:1 to 1:1.
- 29. (previously presented) A polyisobutene as claimed in claim 27, wherein the molar ratio of the alcohol (a) to the ether (b) is from 0.4:1 to 1:1.
- 30. (previously presented) A polyisobutene as claimed in claim 29, wherein the alcohol (a) is isopropyl alcohol and/or 2-butanol.
- 31. (previously presented) A polyisobutene as claimed in claim 29, wherein the ether (b) is diisopropyl ether, di-sec-butyl ether and/or isopropyl sec-butyl ether.
- 32. (previously presented) A polyisobutene as claimed in claim 27, wherein the alcohol (a) is isopropyl alcohol and/or 2-butanol.
- 33. (previously presented) A polyisobutene as claimed in claim 27, wherein the ether (b) is diisopropyl ether, di-sec-butyl ether and/or isopropyl sec-butyl ether.
- 34. (previously presented) A process as claimed in claim 12, wherein the terminal vinylidene group content is

of more than 90%.

35. (previously presented) A process as claimed in claim 15, wherein the terminal vinylidene group content is of more than 90%.

Evidence appendix

Declaration under 37 C.F.R. §1.132 of Dr. Hans-Peter Rath, filed January 16, 2003.

Related proceedings appendix

None.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of	)	
RATH et al.	)	
	)	Art Unit:1713
	)	
Serial No. 09/701,587	)	Examiner: Lu
	)	
Filed: November 30, 2000	)	
	)	
For: PREPARATION OF HIGHLY REACTIVE POLYISOBUTENES		

DECLARATION UNDER 37 CFR §1.132

I, Hans-Peter Rath, Dr., a citizen of the Federal Republic of Germany and residing at Friedhofstrasse 7, 67269 Grünstadt, Germany, declare as follows:

I am a fully trained chemist, having studied chemistry at Technische Universität Aachen (Technical University of Aachen), Germany, during the period of 1965-1974.

I obtained my doctor's degree from Technische Universität Aachen in 1974.

Since 1975, when I joined BASF Aktiengesellschaft of 67056 Ludwigshafen, Germany, I have been working primarily in the field of cationic polymerization.

Therefore, I am familiar with the field to which the subject application relates. I am also familiar with the examiner's rejections of the claims of the subject application.

In the present Declaration, I state the results of Reactivity Tests comparing that of the instant invention with that of a comparative Example.

As outlined in the table below, the following process according to the instant invention and according to US 5,408,018, were compared. Example A as outlined in the table below corresponds to Example 7 of the instant application on page 13, lines 42-45 and in the table on page 14. The example A, corresponding to example 7 of the

application was carried out as follows.

Applicants utilized a recycle reactor consisting of a Teflon tube which had a length of 7.1 m and an internal diameter of 6 mm via which 100 l/h of the reactor content were circulated by means of a gear pump. The tube and pump had a capacity of 200 ml. The Teflon tube and the pump head were immersed in a cooling bath having a temperature of  $-23.8^{\circ}\text{C}$  (cryostat). A mixture of 300 g/h of isobutene and 300 g/h of hexane was dried over a  $3\text{\AA}$  molecule sieve to a water content of less than 3 ppm, and then fed to the recycle reactor through a capillary which had an internal diameter of 2 mm and which was pre-cooled to  $-23^{\circ}\text{C}$ .  $\text{BF}_3$  and isopropyl/diisopropylether as complexing agents were directly introduced into the hexane feed to the reactor. The feed of 10 mmol/h isopropanol and 25 ml/h di-sec-butyl ether and n-hexane (as diluent) was set to 15:1:4, and the amount of  $\text{BF}_3$  was varied until a concentration of isobutene of 1% by weight was obtained.

At a reactor temperature of  $-18^{\circ}\text{C}$  the  $\text{BF}_3$  feed amounted to 21.4 mmol/h. The reactor effluent was washed with water and worked up by distillation at  $230^{\circ}\text{C}/2\text{mbar}$ . The molecular weight  $M_n$  of the polymer was 890, the reactivity (vinylidene double bond content) was 95.9%.

This process was repeated in the comparative example utilizing isopropyl tert-butyl ether, as disclosed in US 5,408,018, was used instead of di-sec-butyl ether. To arrive at a molecular weight in the magnitude obtained in Example A, the amounts of  $\text{BF}_3$  and isopropanol had to be increased over Example A. Thus, as you can see from the results displayed in table form below, the instant process provides an unexpected increase in reactivity over that of the process utilizing isopropyl tert-butyl ether.

The following example shows the results of these tests:

Example	BF <sub>3</sub> [mmol/h]	Isopropanol [mmol/h]	Ether [mmol/h]	Molar Ratios		[Isobutene] weight %	M <sub>n</sub>	Reactivity [%]
				B:I	(I+B):S			
A	21.4	10.0	25.0 Di-sec- butylether	2.5	1.64	1.0	890	95.9
compara tive example	29.7	15.0	25.0 Isopropyl- tert- butylether	2.5	1.36	1.0	960	85.4

Signed at 67056 Ludwigshafen, Germany, this 1. day of Jan, 2003

  
Hans-Peter Rath